Observations of condensed-phase HNO_3 in a tropical subvisual cirrus cloud

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Introduction

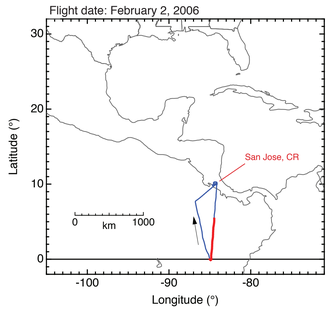
Subvisual cirrus clouds that form near the tropical tropopause have signficant potential for controlling water vapor concentrations in air entering the stratosphere, and they also participate in Earth's radiation budget.

Observations in a subvisual cirrus cloud during the Costa Rica Aura Validation Experiment (CR-AVE) on 2 February 2006 show the presence of condensed-phase HNO_3. This cloud was observed over a broad geographic extent (>700 km) on the southbound leg of a NASA WB-57F flight from San Jose, Costa Rica (shown by the red highlighted region on the flight-track below).

Understanding the role of HNO_3 in the formation and growth of cirrus ice particles may be essential in GCMs for correctly simulating the contribution of subvisual cirrus clouds to the radiative forcing of climate.

NASA WB-57F high-altitude aircraft

Flight date: February 2, 2006



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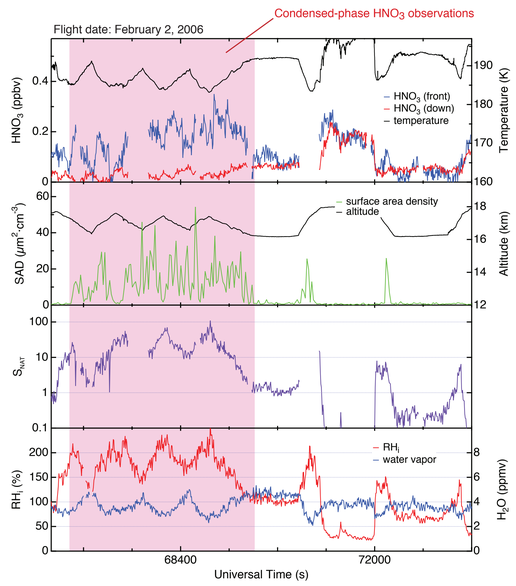


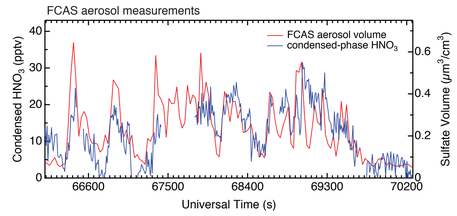
Nitric acid was measured onboard the NASA WB-57F with the NOAA Chemical Ionization Mass Spectrometer (CIMS), which has two independent channels of detection connected to separate foward- and downward-facing inlets (top right). Condensed-phase HNO_3 is revealed by a signicant difference in the mixing ratios observed by the two channels (pink highlighted region, left). The presence of a subvisual cirrus cloud is indicated by enhanced values in the ice particle surface area density (SAD).

This cloud was observed at altitudes of 16-17 km in an extremely cold air mass (183-190 K). Ambient water vapor measurements show that relative humidities with respect to ice ranged from 100-240%. Despite gas-phase HNO_3 mixing ratios of less than 100 pptv, the cold ambient temperatures caused nitric acid trihydrate (NAT) saturation ratios to be 10 or greater during much of the cloud encounter.

Condensed-phase HNO_3 equivalent mixing ratios, calculated by accounting for the particle sampling efficiency of the CIMS inlets, correlate well with measurements of aerosol volume in the cloud (bottom right). Aerosol volume is measured by the Focused Cavity Aerosol Spectrometer (FCAS). The enhanced values of aerosol volume measured in the cloud (>0.1 $\mu m^3 \cdot cm^3$) are assumed to result from the remnants of cirrus ice particles shattered by the FCAS sampling inlet. This result provide additional independent evidence of condensed-phase HNO_3.

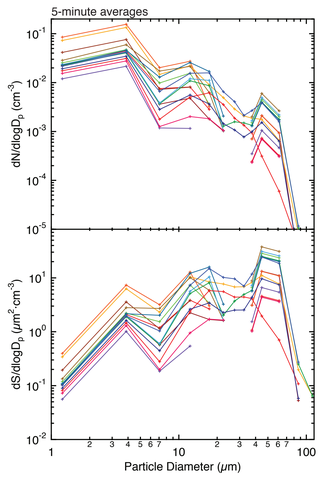
NOAA CIMS inlet pylon

FCAS aerosol measurements



Ice particle measurements

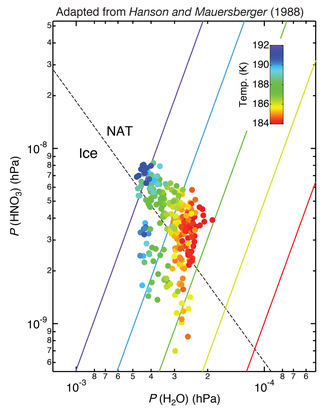
Measurements of ice particle concentration with the Cloud Aerosol and Precipitation Spectrometer (CAPS) indicate the presence of large ice crystals in this cloud (50-100 μm), as well as a smaller particle mode in the 10-20 μm range. Ice particle surface area densities in the cloud were typically less than 50 $\mu m^2 \cdot cm^{-3}$. The surface are density is dominated by the large particle mode (bottom panel).



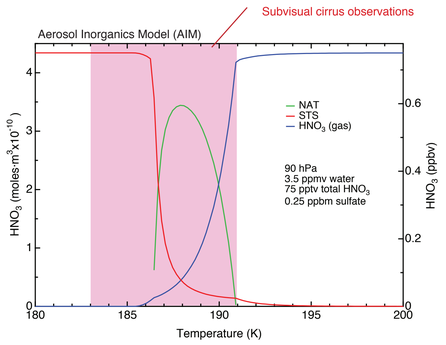
Particle composition

Simultaneous measurements of total HNO_3, water vapor, and temperature indicate that much of the cloud is saturated with respect to nitric acid trihydrate (NAT, below left). This result raises the possibility that HNO_3 may be present in the cloud as a stable condensate, and not simply physically adsorbed on (or trapped in) the ice particles. Results from the Aerosol Inorganics Model (AIM) indicate that the background sulfate aerosol will deliquesce and take up HNO_3 as a supercooled ternary solution (STS) at temperatures below approximately 187 K (below right). Neither the phase equilibria described by *Hanson and Mauersberger* (1988) or the AIM, however, consider the physical adsorption of HNO_3 on cirrus ice particles.

Adapted from *Hanson and Mauersberger* (1988)



Aerosol Inorganics Model (AIM)



Conclusions and implications

Condensed-phase HNO_3 was observed in a tropical subvisual cirrus cloud during the Costa Rica Aura Valication Experiment.

Thermodynamic considerations suggest the condensed-phase HNO_3 may exist as NAT or STS, or be physically adsorbed on the ice particles.

Satellite retrievals of HNO_3 mixing ratio near the tropical tropopause need to account for the sequestration of HNO_3 in the condensed phase when subvisual cirrus clouds are present.

Future measurements near the tropical tropopause will allow us to further characterize the distribution of HNO_3 in low-temperature cirrus clouds and assess the role of HNO_3 in the formation and growth of cirrus ice particles.

Thanks to the air and ground crews of the NASA WB-57 aircraft at Ellington Field, Houston.